Liquid-phase epitaxial growth of very thick In1-xGaxAs layers with uniform composition by source-current-controlled method

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Liquid-phase epitaxial growth of very thick $In_{1-x}Ga_xAs$ layers with uniform composition by source-current-controlled method

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A source-current-controlled method for the continuous supply of solute elements into solutions during growth was developed. In this method, a continuous electric current is passed through a GaAs source material of the solute elements during growth and the source material is dissolved into the growth solution due to Peltier and Joule heating at the interface between the source material and the solution. This method was applied to the liquid-phase epitaxial growth of $In_{1-x}Ga_xAs$ on InP. The temperature variation of the solution was directly measured by immersing a thermocouple in the solution and a temperature gradient of more than 6 °C could easily be obtained in the solution between the GaAs source material and the InP substrate. The amount of dissolved GaAs was measured as a function of the current density. Using these results, an optimum growth condition for the continuous supply of solute elements was determined, and an 80-μm-thick $In_{0.54}Ga_{0.46}As$ layer with uniform composition ($x = 0.458 \pm 0.002$) was obtained.

I. INTRODUCTION

Epitaxial layers with good quality, such as $In_{0.53}Ga_{0.47}As$, $In_{1-x}Ga_xAs_{1-y}P_y$, and $In_{0.49}Ga_{0.51}P$ compounds, must be lattice matched to substrates of binary compounds. Although there are many useful compounds, such as $GaAs_xP_{1-x}$ ($x \approx 0.5$) for short wavelength lasers and $In_{1-x}Ga_xAs$ ($x > 0.5$) for effective carrier confinement layers of long wavelength lasers, these compounds are not available because there are no suitable substrates. To use these compounds, homogeneous ternary bulk crystals are necessary as substrates. The toughest problem in the preparation of homogeneous ternary bulk crystals is variation in the composition of the crystals due to depletion of solute elements during growth. It is also very difficult to grow more than 5-μm-thick epitaxial layers with uniform composition by the conventional liquid-phase epitaxial (LPE) method.

To solve the problem of solute element depletion, Daniele *et al.*[1–5] developed a unique method to control the supply of solute elements to the growth solution. They used Peltier-induced LPE[1,2] to grow $Al_xGa_{1-x}As$ (Refs. 3 and 4) and $In_{1-x}Ga_xP$ (Ref. 5) thick layers of constant composition. In their method, an electric current was passed through the substrate-solution interface, and a temperature gradient was generated in the solution due to the Peltier cooling. A thick crust of AlGaAs or InGaP was floated on the surface of the solution. Solute elements were supplied from the crust to the substrate by diffusion due to the temperature gradient.

Nakajima *et al.*[6–8] have developed the source-current-controlled (SCC) method to control the supply of solute elements to growth solutions during growth. In their method, a pulsed electric current was directly passed through a binary semiconductor compound (GaAs or InAs) used as a source material of the solute elements (Ga, As, or In). The source compound was dissolved into the saturated growth solution due to Peltier and Joule heating at the interface between the source compound and the growth solution. Thus, the source compound played a dual role as the source material for solute elements and as the self-heater in the SCC method. An electric current was not intentionally passed through the substrate-solution interface in order to prevent dissolution of the substrate by Joule heating. In their experiments, the SCC method was applied to the LPE growth of $In_{1-x}Ga_xAs$ on InP. It was found that enough solute elements to adjust the compositional variation in LPE layers could be supplied to the growth solutions during growth by the electric current passed through the source compounds. Very thick ($\sim 200\ \mu$m) $In_{1-x}Ga_xAs$ LPE layers could be grown on InP while passing a pulsed electric current through a GaAs source compound; these LPE layers did not have a uniform composition but, rather, showed compositional variation.

In this work, the SCC method, in which a direct electric current was continuously passed through a GaAs source compound during growth, was first applied to the LPE growth of $In_{1-x}Ga_xAs$ on InP in order to determine if very thick $In_{1-x}Ga_xAs$ layers with uniform composition could be obtained. The compositional variation in these thick LPE layers was measured by electron-probe microanalysis (EPMA). To determine whether the temperature gradient in the solution could be controlled by electric current, the temperature variation of the In-Ga-As solution was directly measured by immersing a thermocouple in the solution while passing a current through the GaAs source compound. The amount of dissolved GaAs source compound was measured as a function of the current density and the temperature of the solution to determine whether the supply of solute elements from the source compound could be controlled by electric current. Using these results, an optimum current density to obtain very thick $In_{1-x}Ga_xAs$ LPE layers with uniform composition by the continuous supply of solute elements was determined.

II. EXPERIMENTAL PROCEDURE AND RESULTS

A. LPE growth conditions for lattice matching

The liquidus data of the In-Ga-As system near 840 °C (Refs. 9–11) were used to grow $In_{1-x}Ga_xAs$ on InP

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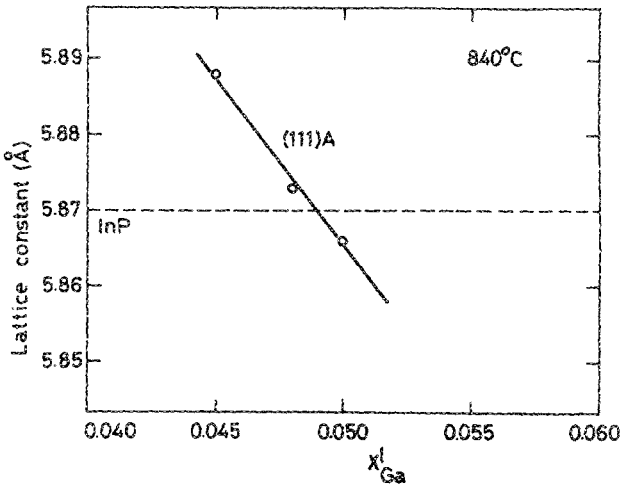

FIG. 1. Lattice constant of (111)A $In_{1-x}Ga_xAs$ LPE layers displayed as a function of X^l_{Ga}.

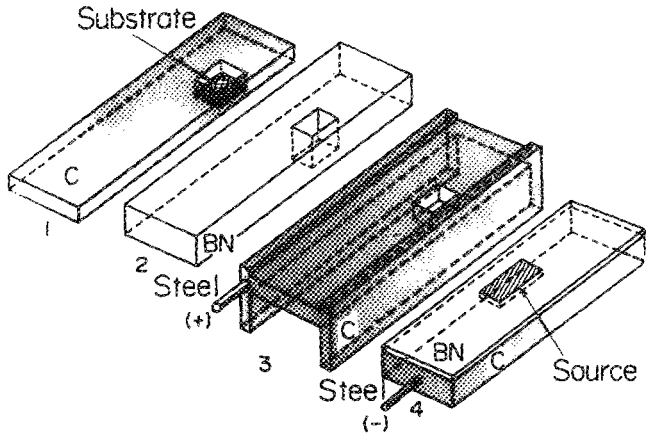

FIG. 2. Schematic drawing of the parts of the boat used in this work.

(111)A substrates by the LPE method. The LPE growth conditions for lattice-matched $In_{0.53}Ga_{0.47}As$ layers on InP (111)A substrates were found by lattice constant measurements of the ternary LPE layers grown from solutions with compositions on the liquidus isotherm at 840 °C. The LPE growth apparatus consisted of a horizontal furnace system and a conventional sliding graphite boat, as previously reported,[12] and semiconductor-grade materials were used.

Just prior to loading, the substrates were etched in (90 H_2SO_4/5 H_2O_2/5 H_2O) for about 3 min. Prior to growth, the substrates were cleaned by a meltback technique using undersaturated In-P solutions. $In_{1-x}Ga_xAs$ layers were grown starting from 840 to 839 °C at a constant cooling rate of 1 °C/min by the ramp-cooling method. No supersaturated solutions were used in the growth. The thickness of the LPE layers was 5–6 μm. The lattice constants of $In_{1-x}Ga_xAs$ layers were measured by the double-crystal x-ray diffraction technique.[13] The precise diffraction angles of the ternary layers were determined from the (333) Cu$K\alpha_1$ reflection by using the substrate reflection as an internal standard.

The lattice constant is displayed as a function of X^l_{Ga} in Fig. 1, where X^l_i represents the atomic fraction of an element i in the In-Ga-As ternary solution. The lattice constant is of the epitaxial lattice perpendicular to the wafer surface. The dashed line in Fig. 1 represents the lattice constant of InP. The solution composition required to grow a lattice-matched layer on (111)A InP is $X^l_{Ga} = 0.049$, $X^l_{As} = 0.213$, and $X^l_{In} = 0.738$ and it is just saturated at 840 °C. The distribution coefficients of Ga and As on the (111)A face at 840 °C are 4.8 and 2.3, respectively. The distribution coefficients of Ga and As on the (111)A face at 650 °C are 9.2 and 8.5, respectively.[14] The distribution coefficients at 840 °C are smaller than those at 650 °C.

B. Apparatus and principle of the SCC method

Figure 2 shows a schematic drawing of the parts of the boat used in this work. In this drawing, BN represents boron nitride used as insulation, and C represents carbon. The boat consists of four parts, 1, 2, 3, and 4. Part 1 is a carbon boat slider with a well. A substrate is set in the well. Part 2 is a boron nitride block with a well in which a solution is put. Part 3 is a carbon boat with an extension of the well in part 2. This boat has a positive electrode (+) made of stainless steel. Part 4 is a carbon-boat holder with a boron nitride plate and a negative electrode (−) made of stainless steel. A source compound is set in a pit through this boron nitride plate. The electrodes are screwed into the carbon boats. These parts (1, 2, 3, and 4) were assembled into a boat, as shown in Fig. 3. Cross sections of this boat are shown in Fig. 4. When the In-Ga-As solution is heated before growth, the solution and parts of the boat are arranged as shown in Fig. 4-1. The solution is not in contact with either the substrate or the source compound. After heating, when an $In_{1-x}Ga_xAs$ epitaxial layer is being grown, the InP substrate, the In-Ga-As solution, and the GaAs source compound are arranged as shown in Fig. 4-2. The substrate is on the top of the solution. A part of the surface of the source compound is in contact with the bottom of the solution and the opposite side touches the carbon-boat holder through an In contact melt. The solution is surrounded by boron nitride, but only the upper

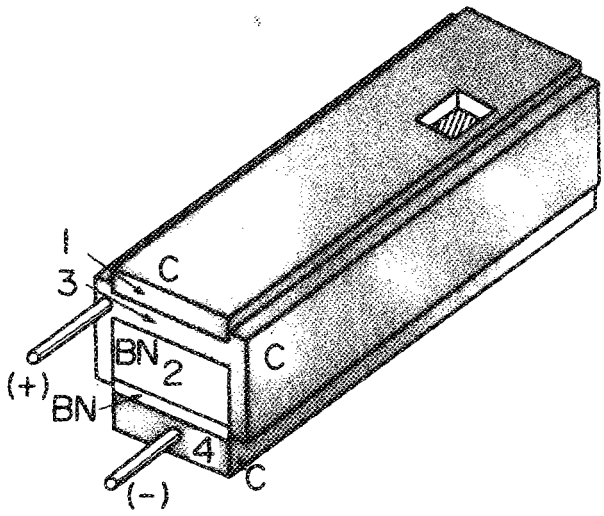

FIG. 3. Schematic drawing of the boat used in this work.

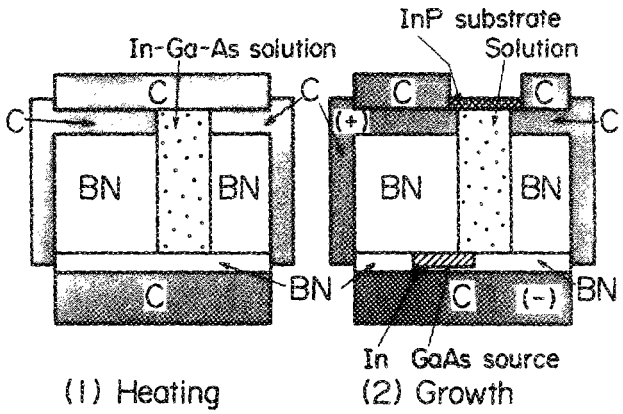

FIG. 4. Cross sections of the boat when the In-Ga-As solution is heated before growth (1) and when an $In_{1-x}Ga_xAs$ LPE layer is grown (2).

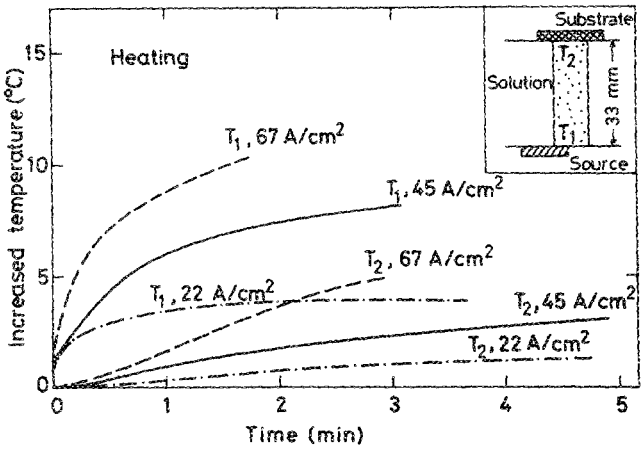

FIG. 6. Increased temperature from 840 °C in the solution as a function of the length of time in which current was passed through the source compound.

part of the solution touches carbon. The depth of the solution was 33 mm.

A direct electric current is continuously passed through the carbon boat, the solution, the source compound, and the carbon-boat holder, in that order. Figure 5 explains the principle of the SCC method. The temperature, T_1, just above the source compound increases due to Peltier heating and Joule heating. The temperature T_1 is always higher than the temperature T_2 just below the substrate. Thus, a temperature gradient is generated in the solution between the source compound and the substrate. The movement of solute elements from the bottom to the top is promoted by diffusion due to the temperature gradient. The heat dissolves the source compound and continuously supplies solute elements to the solution. Thus, the $In_{1-x}Ga_xAs$ LPE layer can be grown while solute elements are being continuously supplied by dissolution of the source compound and by diffusion due to the temperature gradient.

C. Measurements of temperature gradient in solution

The temperature variation in the In-Ga-As solution was directly measured by immersing a thermocouple in the solution while passing electric current through the GaAs source compound, in order to determine whether the temperature gradient in the solution could be controlled by electric current. The thermocouple was protected by a fine silica tube in the solution. The ternary solution was just saturated at 840 °C and the depth of the solution was 33 mm. After the temperature of the solution was kept constant at 840 °C, an electric current was passed to increase the temperature. Figure 6 shows the increased temperature from 840 °C in the solution, as a function of the length of time in which current was passed through the source compound. The temperature was monitored at two points in the solution. T_1 is the temperature near the source compound and T_2 is the temperature at the top of the solution. When a current of 45 A/cm^2 was passed through the source compound, temperature T_1 rapidly increased and temperature T_2 gradually increased. Both temperatures eventually saturated as the length of time increased. A temperature gradient of more than 5 °C could be obtained. When a current of 67 A/cm^2 was passed through the source compound, temperatures T_1 and T_2 increased more rapidly. A temperature gradient of more than 7 °C could be obtained. When a current of 22 A/cm^2 was passed, temperature T_1 increased rapidly only at the beginning and became saturated in a short time. Temperature T_2 increased by only 1.3 °C. As the current density decreased,

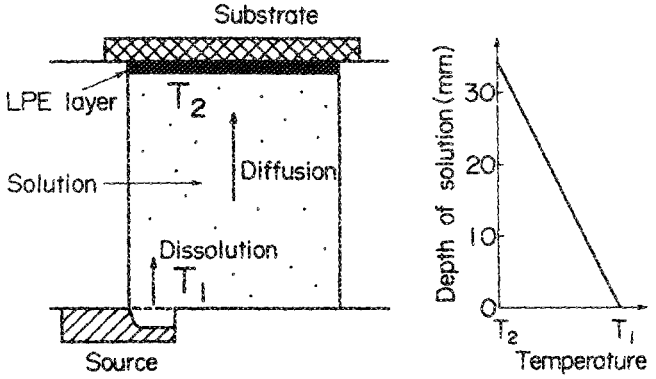

FIG. 5. Principle of the SCC method.

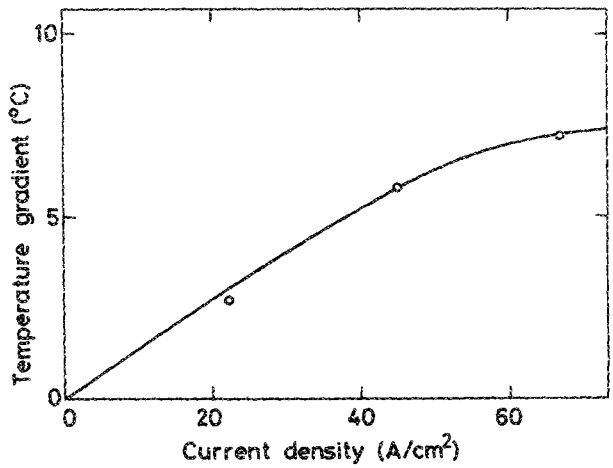

FIG. 7. Temperature gradient, $T_1 - T_2$, as a function of the current density.

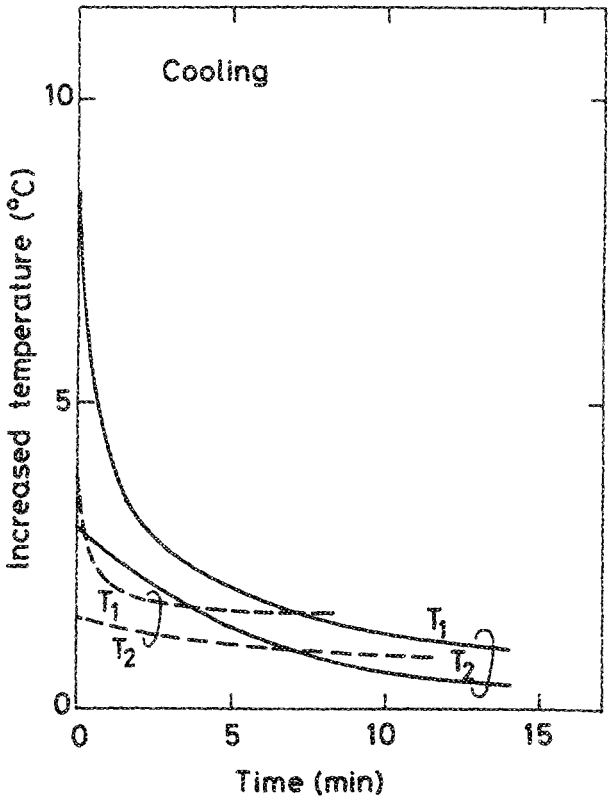

FIG. 8. Cooling curve of the increased temperature after the current was cut off.

both temperatures became saturated in a shorter time. Figure 7 shows the temperature gradient, $T_1 - T_2$, as a function of the current density. It confirms that the temperature gradient can be controlled only by electric current. Figure 8 shows how the increased temperature was cooled after the current was cut off. As the increased temperature was higher, the temperature was cooled more rapidly. However, it was very difficult for the temperature to be cooled to the initial temperature of 840 °C within 15 min because the heat capacity of the carbon boat was fairly large.

D. Measurements of the amount of dissolved GaAs source compound

To determine whether the supply of solute elements from the source compound could be controlled by electric current, the amount of the dissolved GaAs source compound was measured as a function of the current density, and is shown in Fig. 9. The vertical axis shows the amount of dissolved GaAs source compound per 1 g In-Ga-As solution, whose compositions are $X^l_{Ga} = 0.04$, $X^l_{As} = 0.170$, and $X^l_{In} = 0.790$ at 790 °C and $X^l_{Ga} = 0.05$, $X^l_{As} = 0.212$, and $X^l_{In} = 0.738$ at 840 °C. For each measurement, current flowed for 2 min. There are three parameters which affect dissolution of the source compound. They are the current density, the temperature of the solution, and the direction of the current which controls the presence of Peltier heating. At 790 °C, 260 A/cm^2 is required to dissolve 5 mg of GaAs by Joule heating only. In this case, the current flows from the GaAs source compound to the solution. Only Joule heating occurs and Peltier cooling also occurs at the interface

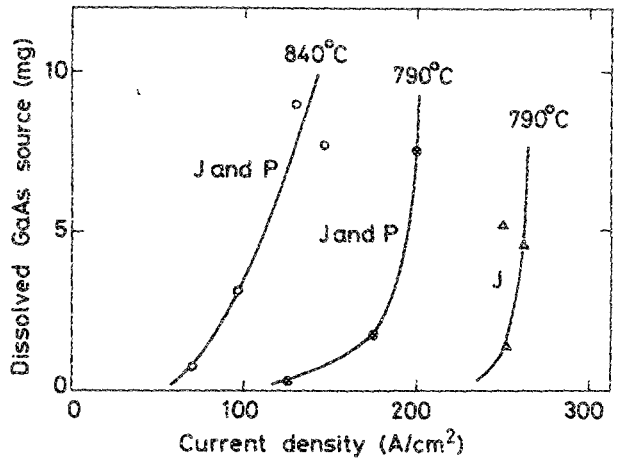

FIG. 9. Amount of dissolved GaAs source compound as a function of the current density.

between the source compound and the solution. When the current flows from the solution to the source compound for the purpose of using both Peltier and Joule heating, the current density required to dissolve 5 mg of GaAs can be reduced to 195 A/cm^2. The current density required to dissolved the same amount of GaAs at 840 °C is about half that at 790 °C. The GaAs source compound abruptly dissolves at a threshold current density because the heat which contributes to dissolution is superior to the heat which scatters through the carbon boat at the threshold point. As shown in Fig. 9, the supply of solute elements from the GaAs source compound can be controlled by the electric current.

E. LPE growth of very thick layers with uniform composition by the SCC method

To determine whether very thick epitaxial layers with uniform composition could be obtained by the SCC method, this method was first applied to the LPE growth of $In_{1-x}Ga_xAs$ on InP.

Figure 10 is a temperature diagram at the top of the solution. The solution, without contact with either the sub-

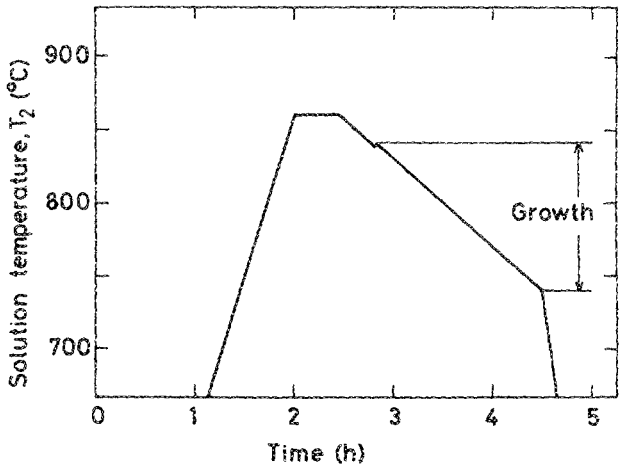

FIG. 10. Temperature diagram of the top of the solution for the growth of very thick $In_{1-x}Ga_xAs$ LPE layers by the SCC method.

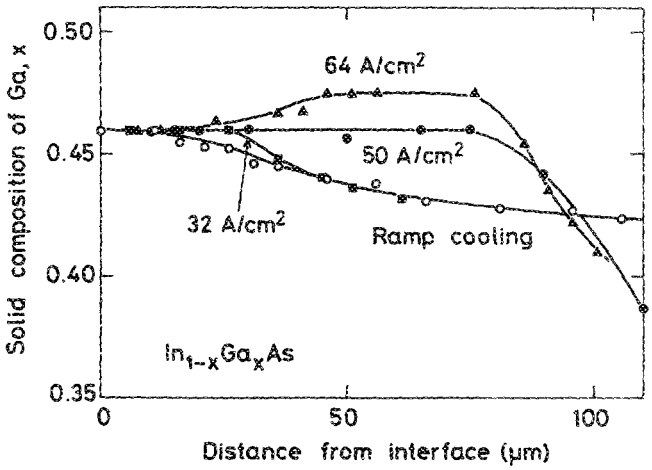

FIG. 11. Solid composition of Ga, x, in very thick $In_{1-x}Ga_xAs$ LPE layers as a function of distance from the interface between the layers and InP substrates.

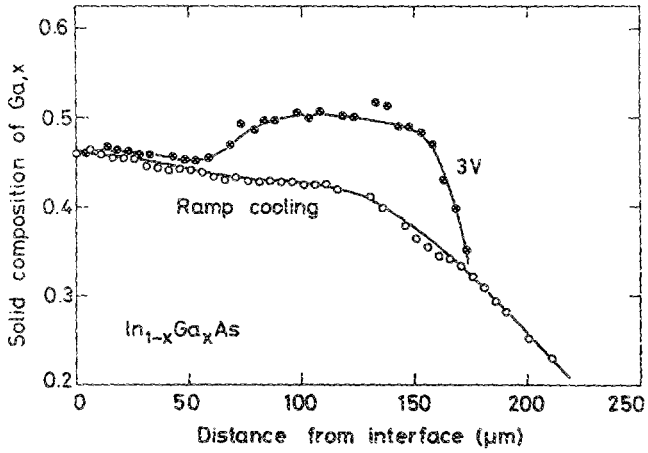

FIG. 12. Solid composition of Ga, x, in an $In_{1-x}Ga_xAs$ LPE layers grown by the SCC method applying a constant voltage of 3 V, together with the solid composition in the LPE layer grown by only conventional ramp cooling.

strate or the source compound, was rapidly heated to 860 °C. This is 20 °C above the saturation temperature of the solution. The solution was held at 860 °C for about 30 min. Then, the solution was cooled to 840 °C at a constant cooling rate of 1 °C/min, and was brought in contact with the substrate and the source compound. At this temperature, the electric current was continuously passed through the GaAs source compound. The temperature of the solution increased, and a temperature gradient was generated in the solution. The GaAs source compound was continuously dissolved in the solution. An $In_{1-x}Ga_xAs$ LPE layer was grown on (111)A InP from 840 to 780 or 740 °C while solute elements were being continuously supplied by dissolution and diffusion.

Figure 11 shows the solid composition of Ga, x, in the $In_{1-x}Ga_xAs$ layers as a function of the distance from the interface between the layers and the InP substrates. The open circles show the composition of a layer grown only by conventional ramp cooling from 840 to 780 °C. A constant cooling rate of 1 °C/min was used. The composition varies from 0.46 at the interface to 0.42 at the surface due to depletion of Ga in the solution. The solid squares show the composition of a layer grown from a solution in which solute Ga and As were supplied by a current of 32 A/cm^2. The composition variation was partially compensated. When a current of 64 A/cm^2 was passed through the GaAs source compound, the composition of Ga increased to more than 0.46 because of the oversupply of solute elements, as shown by the solid triangles. These two layers of 32 and 64 A/cm^2 were grown through cooling from 840 to 740 °C. When a current of 50 A/cm^2 continuously flowed, an approximately 80-μm-thick $In_{0.54}Ga_{0.46}As$ LPE layer with extremely uniform composition ($x = 0.458 \pm 0.002$) was obtained for the first time, as shown by the solid dots. This layer was grown by cooling from 840 to 780 °C. Under this optimum growth condition, solute elements were successfully supplied by dissolution and diffusion. The solid compositions of the two layers grown with 50 and 64 A/cm^2, shown by the solid dots and the solid triangles, rapidly decrease when the currents were cut off and the solutions were rapidly cooled to room temperature, because the solutions could not be removed from the surfaces of the layers when the growth was terminated.

Under the condition of a constant flowing current, the heating value decreases with dissolution of the source compound because its resistivity decreases. Under the condition of a constant applied voltage, however, the heating value does not decrease because the flowing current increases as the resistivity decreases. Figure 12 shows the solid composition x of an $In_{1-x}Ga_xAs$ LPE layer grown by the SCC method applying a constant voltage of 3 V, as shown by the solid dots. The composition is represented as a function of the distance from the interface between the layer and the InP substrate. The LPE layer was grown by cooling from 840 to 740 °C. The current density increases from 39 A/cm^2 at the start of the run to 99 A/cm^2 at the finish. Therefore, the solid composition significantly increases, starting at the midpoint of the growth run. This indicates the oversupply of solute

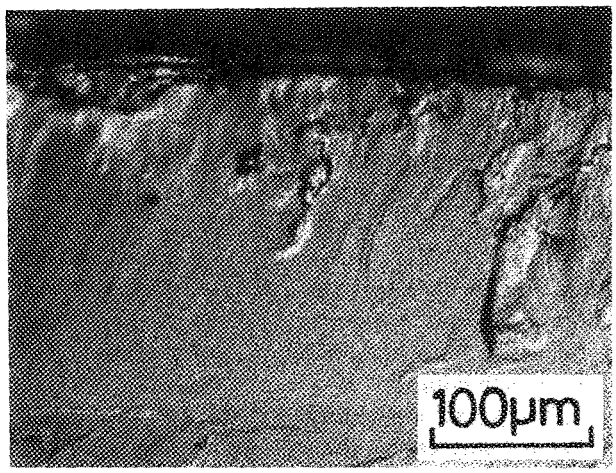

FIG. 13. Cross section of an $In_{1-x}Ga_xAs$ layer grown by the SCC method.

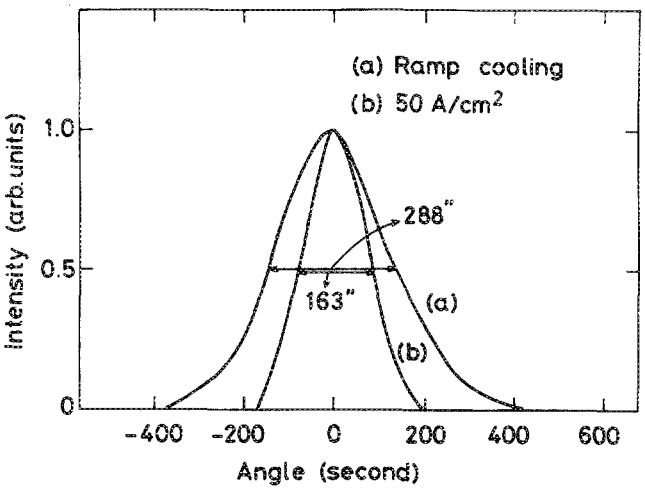

FIG. 14. Cu$K\alpha_1$ rocking curves for the (333) reflection of very thick $In_{1-x}Ga_xAs$ LPE layers grown on InP (111)A substrates.

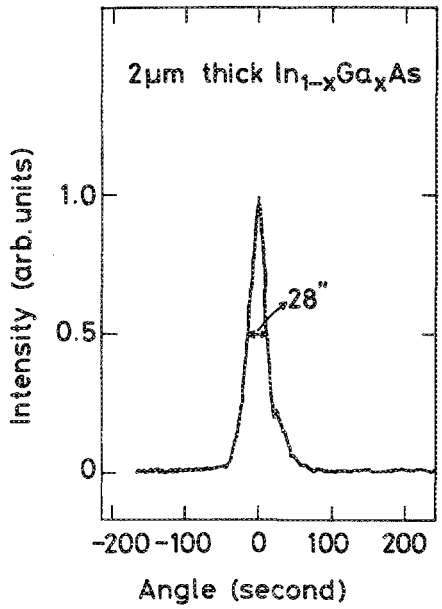

FIG. 15. Cu$K\alpha_1$ rocking curves for the (333) reflection of a lattice-matched $In_{0.53}Ga_{0.47}As$ LPE layer grown on an InP (111)A substrate.

elements from the source compound. The open circles show the composition of a layer grown by only conventional ramp cooling from 840 to 780 °C, at a constant cooling rate of 1 °C/min. From Figs. 11 and 12, it is required that the heating value gradually decreases as the growth advances, to obtain a layer with uniform composition.

Figure 13 shows the cross section of an $In_{1-x}Ga_xAs$ epitaxial layer grown by the SCC method. The thickness of the layer is 210 μm.

F. X-ray rocking curves of thick layers

Figure 14 shows the Cu$K\alpha_1$ rocking curves for the (333) reflection of very thick $In_{1-x}Ga_xAs$ LPE layers grown on InP (111)A substrates. Curve (a) is the x-ray rocking curve of the layer grown by only ramp cooling, which is shown in Figs. 11 and 12. Curve (b) is for a layer grown from a solution in which solute elements were supplied by a current of 50 A/cm^2, which is shown in Fig. 11. The half-width of the diffraction peak of curve (b) (about 163 s) is much smaller than that of curve (a) (about 288 s). This indicates that the composition variation of the LPE layers of curve (b) is much smaller than that of curve (a). The typical half-width of the diffraction peak of lattice-matched $In_{0.53}Ga_{0.47}As$ on (111)A InP is less than 30 s, as shown in Fig. 15. Figure 15 is the Cu$K\alpha_1$ rocking curve for the (333) reflection of this layer and its thickness is 2 μm. These results indicate that large stress still remains in the very thick LPE layer because the part of the layer grown after the current was cut off has large compositional variation, as shown in Fig. 11.

G. Relation between layer thickness and current density

Figure 16 shows the thickness of $In_{1-x}Ga_xAs$ LPE layers grown by the SCC method as a function of the current density (A/cm^2) flowing through GaAs source compounds. These layers were grown by cooling from 840 to 740 °C. The layer thickness gradually increases as the current density increases. This indicates that the amount of supplied solute elements increases as the current density increases. The layer thickness eventually saturates because the temperature gradients also saturate as the current density increases, as shown in Fig. 7.

III. CALCULATION

A. Calculation of compositional variation in thick layers

An expression to calculate the layer thickness is derived on the basis of the assumption that the solution is always uniform throughout its volume during the growth.[15,16] The amount of solid deposited from the uniform solution during the cooling cycle from T to $T-\Delta T$ can be estimated by the lever rule[17] as follows:

$$\frac{n-s}{s}=\frac{0.5-X^l_{As}}{X^l_{As}-(X^l_{As}-\Delta X^l_{As})}, \tag{1}$$

where X^l_{As} is the atomic fraction of As in the In-Ga-As ter-

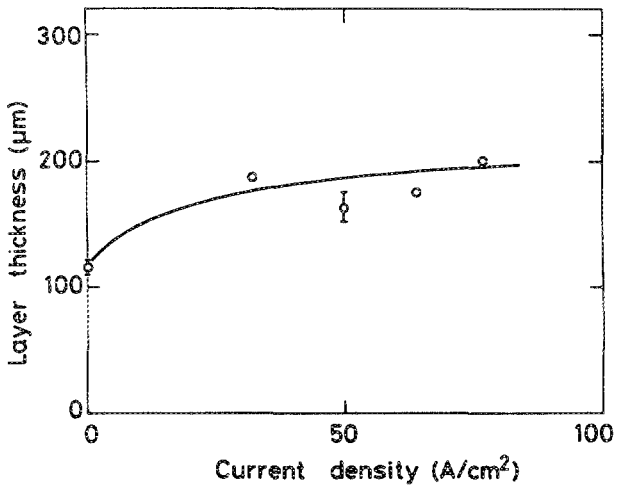

FIG. 16. Thickness of $In_{1-x}Ga_xAs$ LPE layers grown by the SCC method as a function of the current density (A/cm^2) flowing through GaAs source compounds.

TABLE I. Thermodynamic input data for the calculation of compositional variation in $In_{1-x}Ga_xAs$ thick layers.

Input data	Ref.	Input data	Ref.
$T^F_{GaAs} = 1511$ K	19	$T^F_{InAs} = 1215$ K	22
$\Delta S^F_{GaAs} = 16.64$ cal/mol K	20	$\Delta S^F_{InAs} = 14.52$ cal/mol K	20
$\Omega^l_{GaAs} = 5160 - 9.16T$ cal/mol	21	$\Omega^l_{InAs} = 3860 - 10.0T$ cal/mol	23
$\Omega^l_{GaIn} = 2300$ cal/mol		$\Omega^s_{GaAs\text{-}InAs} = 3300$ cal/mol	

nary solution at a starting growth temperature T, ΔX^l_{As} is the minute change of X^l_{As} for minute change of growth temperature from T to $T - \Delta T$, and s is the number of atoms in the deposited solid from the solution in which the total number of atoms is n. From Eq. (1), the layer thickness L is given in the form

$$L = \frac{n}{C_s A} \frac{\Delta X^l_{As}}{0.5 - (X^l_{As} - \Delta X^l_{As})}, \tag{2}$$

where C_s is the number of atoms per unit volume of the deposited solid, and A is the area of the substrate. When the variation of the lattice constant is not negligible, layer thickness for all steps of infinitesimal change of growth temperature ΔT must be calculated by Eq. (2) with the variable C_s along the deposition path,[8,15,16] and they must be added to obtain the total layer thickness. The ratio n/A in Eq. (2) is given by

$$n/A = \rho_m w N / M_m, \tag{3}$$

where ρ_m, w, N, and M_m are the density of the solution, the solution thickness, Avogadro's number ($= 6.02252 \times 10^{23}$/mol) and the atomic weight of the solution element, respectively. In this calculation, ρ_m and M_m are the density of In in the solution at 650 °C ($\rho_m = 6.66$ g/cm^3) (Ref. 18) and the atomic weight of In ($M_m = 114.82$), respectively. In order to calculate C_s for each growth step, the lattice constant of $In_{1-x}Ga_xAs$ layers was calculated on the basis of Vegard's law using the lattice constants of the binary constituent. The procedure described above is repeated until the entire cooling is finished.

The interaction parameters used for the calculation are listed in Table I. In Table I, ΔT^F_{AB} and ΔS^F_{AB} are the temperature and entropy of fusion of the AB compound, respectively. Ω^l and Ω^s are the interaction parameters in the liquid solution and solid, respectively. Two parameters, Ω^l_{GaIn} and $\Omega^s_{GaAs\text{-}InAs}$, were changed to obtain reasonable agreement with the solid composition at 840 °C, whose parameters are similar to those previously reported.[23,24] In this calculation, 0.001 was used as ΔX^l_{As} and the corresponding ΔT was less than 1.5 °C. The starting growth temperature was 840 °C.

Figure 17 shows the calculated solid composition of Ga, x, in $In_{1-x}Ga_xAs$ layers as a function of the distance from the interface between the layers and the substrates for various depths of the In-Ga-As solution. The starting solution composition is $X^l_{Ga} = 0.044$, $X^l_{In} = 0.744$, and $X^l_{As} = 0.212$ at 840 °C. The starting composition of the solid grown from this solution is $x = 0.45$. The broken line shows the solid composition of 0.45. The compositional variation becomes larger as the depth of the solution becomes shallower. When the depth is 1 mm, x varies from 0.45 to 0.30 only at the distance of 65 μm. When the depth is 33 mm, however, x varies from 0.45 to only 0.44, even at a distance of 100 μm.

As compared with Figs. 11 and 17, the compositional variation in the layer grown only by ramp cooling agrees well with the calculated compositional variation of the layer grown from a solution with a depth of 5 mm. This indicates that only 5 mm out of the 33-mm-deep solution is effectively

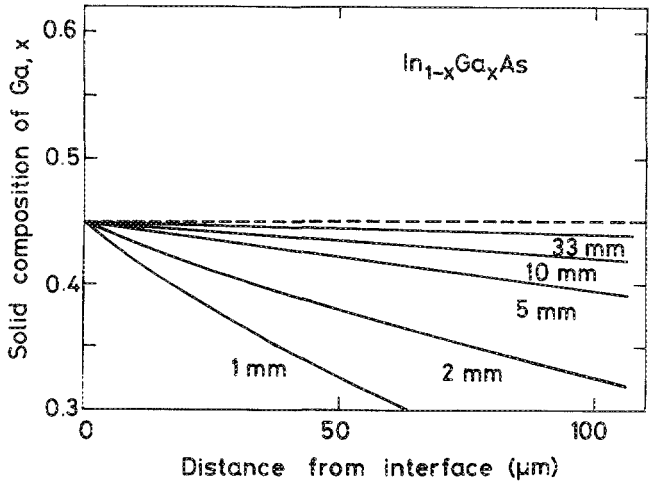

FIG. 17. Calculated solid composition of Ga, x, in $In_{1-x}Ga_xAs$ layers as a function of distance from the interface between the layers and the substrates, for various depths of the In-Ga-As solution.

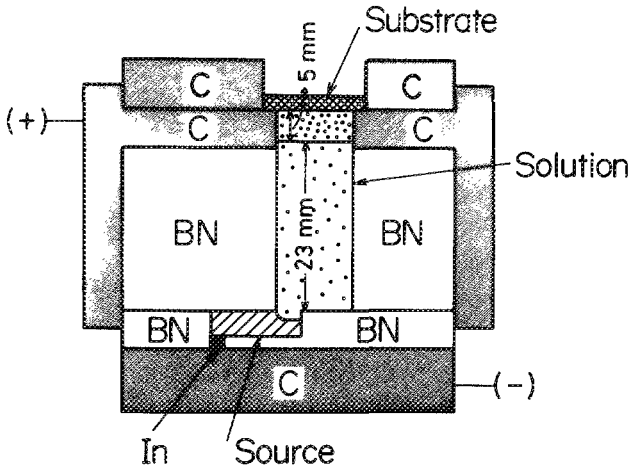

FIG. 18. Cross section of the boat when only 5 mm out of the 33-mm-deep solution is effectively used.

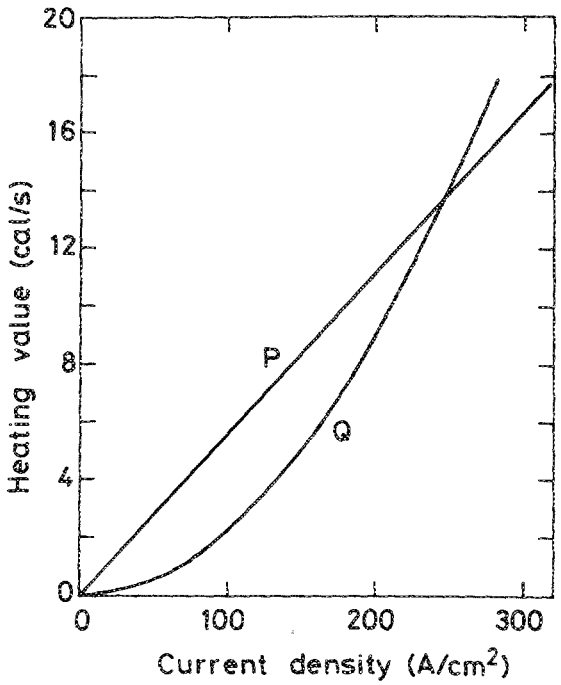

FIG. 19. Estimated Peltier heating value P, and Joule heating value Q, generated in the GaAs source compound as a function of the current density J (A/cm^2).

used, as schematicaliy shown in Fig. 18. When all of the 33-mm-deep solution is effectively used, the compositional variation is fairly constant, as shown in Fig. 17. In order to obtain a layer with uniform composition, a current of 50 A/cm^2 must be passed through the GaAs source compound. In this case, all of the 33-mm-deep solution is effectively used by dissolution of the solute elements due to the temperature gradient, and some solute elements are supplied by dissolution of the source compound.

B. Calculation of Peltier and Joule heating

The magnitude of Peltier and Joule heating in these experiments was estimated. The magnitude of the heat generated by Peltier heating, $P(W)$, is given by[25]

$$P = \pi_p J s, \tag{4}$$

where π_p (V) is the Peltier coefficient, J(A/cm^2) is the density of the electric current, and s(cm^2) is the area of the GaAs source compound in contact with the solution. π_p of n-type GaAs is about 0.4 V near 800 °C (Ref. 20) and s is 0.14 cm^2. The magnitude of the heat generated by Joule heating, Q(W/cm^2), is given by

$$Q = \rho J^2 s l, \tag{5}$$

where ρ (Ω cm) is the resistivity of the source compound and l (cm) is the depth of the source compound. The resistivity ρ of the Si-doped GaAs used in this work is 0.002 Ω cm, and l of the GaAs source compound is 0.8 cm. The Peltier heating value, P, and the Joule heating value Q, were estimated by using Eqs. (4) and (5). The estimated result is shown in Fig. 19. Figure 19 shows the heating value (cal) per 1 s as a function of J(A/cm^2). When J is smaller than 250 A/cm^2, the Peltier heating value P, is larger than the Joule heating value Q. The total heating value of P and Q at $J = 50$ A/cm^2 is about 3.3 cal/s. In order to make P larger than Q, it is necessary to use a source compound with small ρ (Ω cm) and l (cm).

IV. DISCUSSION

These are the preliminary results of the SCC method, but its usefulness is demonstrated by the LPE growth of $In_{1-x}Ga_xAs$ on InP. These results show that enough solute elements to obtain an LPE layer with uniform composition can be supplied into the growth solution by flowing the electric current through the source compound. In this work, the epitaxial layers were grown by the ramp-cooling method using dilute solutions. Thicker LPE layers with uniform composition can be obtained by the SCC method using InAs-GaAs quasibinary melts. In this case, it is able to grow thicker LPE layers at a constant growth temperature while solute elements are being supplied by the dissolution and the diffusion due to the temperature gradient in the solution. The growth of homogeneous ternary bulk crystals by the crystal pulling system needs two essential techniques. One is how to prevent dendritic growth due to constitutional supercooling and another is how to supply depleted solute elements into growth solutions. Thus, homogeneous ternary bulk crystals can be grown combining the SCC method with a method for preventing dendritic growth.

It is important to obtain the ternary seed crystals initially used for the crystal pulling system. These crystals can be obtained from a part of the ternary bulk crystals which are grown using binary seed crystals, such as InP or GaAs, and which have gradual compositional gradient in them. Ternary seed cyrstals with largely different lattice constants from the binary seed crystals can be obtained by repeating the above process again.

V. CONCLUSIONS

The effectiveness of our new method was confirmed by the LPE growth of $In_{1-x}Ga_xAs$. The temperature gradient in the solution can be controlled by the electric current flowing through the source compound due to Peltier and Joule heating. The solute elements are continuously supplied to the solution due to successful dissolution of the source compound by heat. An 80-μm-thick $In_{0.54}Ga_{0.46}As$ LPE layer with uniform composition ($x = 0.458 \pm 0.002$) can be first grown by the SCC method. These results show that applying the SCC method to the crystal pulling system may open the way to obtaining homogeneous ternary bulk crystals.

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